

InGaAs/GaAs Quantum Dots: effects of ensemble interactions, interdiffusion, segregation and proton irradiation.

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A summary of recent experimental findings on the effects of interdiffusion, segregation, strained ensemble interactions and proton irradiation on the optical properties of InGaAs/GaAs quantum dots (QDs) are presented.

Tuning the photoluminescence (PL) emission from QD structures is important for device applications and basic studies. This can be achieved by changing the III-V bandgap (composition) in the dot/barrier materials [1], by adjusting average island dimensions for a given ternary composition [2], and by interdiffusion of the dot/barrier interface [3]. The latter method has recently shown that large blue shifts are obtained with post-growth annealing in InGaAs/AlInAs/GaAs/AlGaAs QDs [4]. Appreciable shifts were also observed after high-temperature growths of upper cladding layers, making this issue relevant to devices.

Values for diffusivities were obtained from shifts in the ground-state photoluminescence (PL) emission in intermixed quantum wells (QWs) by relating such shifts with diffusion lengths. Interdiffusion changes an abrupt interface to a graded interface, if this change in confining potential is included in Schroedinger's wave equation, diffusion lengths can be correlated with measured blueshifts in PL. Values for diffusivities and activation energy for interdiffusion were determined for high-indium-content QW's (In_{0.5}Ga_{0.5}As/GaAs). Quantum-mechanical numerical calculations modeling changes in the quantum-well (QW) confining potential with interdiffusion have been used to obtain values for diffusivities. These showed transient behavior, and activation energies for interdiffusion (3.5 ± 0.3 eV) were found to be similar to values reported for low-indium-content InGaAs/GaAs QW's. In quantum-dot structures, larger blueshifts were obtained than in QW's under similar conditions [5].

Interfacial compositional disordering has also been used in InGaAs/GaAs quantum dots that show level filling (intensity dependent line-shapes due to excited state emission). This technique can tune the intersublevel energy spacings ($\Delta E_{[(i+1) - i]}$) from QDs. Interdiffusion blue-shifted all levels while lowering values for $\Delta E_{[(i+1) - i]}$. PL measurements showed strong emission from excited states for all $\Delta E_{[(i+1) - i]}$ values, which ranged from 53 to 25 meV. As shown in Fig 1, the intersublevel spacings $\Delta E_{[(i+1) - i]}$ were reduced and could be tuned continuously for values of $\Delta E_{[(i+1) - i]}$ greater, similar and lower than the LO phonon energies in InAs and GaAs [6].

The photoluminescence (PL) emission from InGaAs/GaAs quantum-well (QW) and quantum-dot (QD) structures were also compared after controlled irradiation with 1.5-MeV proton fluxes [7]. Results showed a significant enhancement in radiation tolerance with three-dimensional quantum confinement. Some additional radiation-induced changes in photo-carrier recombination from QDs, which include a slight increase in PL emission with low and intermediate proton doses (from 7×10^{11} to 7×10^{12} /cm²), are also examined. Fig. 2 shows an increase in the integrated PL intensity from the QDs after proton irradiation in a low density QD structure which also shows strong WL emission at low temperature. Similar increases were also observed after proton irradiation of high density QD structures. Since no such increase is observed in the QW structures, we attribute this PL enhancement to effects from

three-dimensional quantum confinement. Reduction of the phonon bottleneck by defect assisted phonon emission has been proposed [8] as a mechanism to explain the bright PL emission in QDs. Introduction of deep level defects as those originated from displacement damage might provide additional relaxation paths [9] for thermalization of carriers and therefore increase the luminescence emission. These results show that the luminescence from QDs structures is inherently radiation tolerant due to the effects of three dimensional quantum confinement. An increase in radiation hardness of as much as two orders of magnitude has been obtained by comparisons with quantum wells of the same composition and placed at the same depth in the structure. Additionally, we show that a slight increase in PL emission from InGaAs/GaAs QDs can be observed with low to moderate proton doses.

Large variations in InGaAs quantum dot concentrations were obtained with simultaneous growths on vicinal GaAs [001] substrates with a range in surface step densities. It was found that decreasing dot-dot separation causes blue-shifts in the ground state and excited states photoluminescence (PL) emission, narrows intersublevel transition energies, shortens luminescence decay times for excited states, and increases inhomogeneous PL broadening. Figure 3 shows TEM micrographs with their corresponding low temperature PL and time resolved PL spectra [10].

Some of these results have similarities with data obtained after post-growth annealing experiments, where interfacial compositional disordering of the InGaAs/GaAs interface was used to tune intersublevel energy spacings in QDs. Comparison of the two sets of experimental results, shown in figure 4, can offer some physical insight, indicating that the blue-shifts and narrower intersublevel transitions are a consequence of shallower confinement rather a consequence of electronic coupling. Increasing dot-dot proximity then has the overall effect of making the confining potential shallower. Trends towards decreasing PL decay times are also seen with increasing dot concentration and with increasing eigenstates. Recent measurements of PL decay times confirm that reductions in confining potentials associated with interdiffusion shorten PL lifetimes [11]. Faster excited state PL decay for the large density samples, due to faster interlevel relaxation could be explained with faster thermalization rates due to the energy differences between transitions related to different levels approaching GaAs LO phonon energies as the dot density increases.

The effects of varying the dot-dot separation on their optical properties was further investigated by temperature dependent and time dependent photoluminescence experiments [12]. We report significant differences in the temperature-dependent and time-resolved photoluminescence (PL) from low and high surface density InGaAs/GaAs quantum dots (QD's). QD's in high densities are found to exhibit an Arrhenius dependence of the PL intensity, while low-density (isolated) QD's display more complex temperature-dependent behavior. Some of these differences are presented in Figure 5 (a). The PL temperature dependence of high density QD samples is attributed to carrier thermal emission and recapture into neighboring QD's. Conversely, in low density QD samples, thermal transfer of carriers between neighboring QD's plays no significant role in the PL temperature dependence. The efficiency of carrier transfer into isolated dots is limited by the rate of carrier transport in the InGaAs wetting layer. Time-resolved PL measurements of carrier transfer times in low and high density QD's as a function of sample temperature are shown in 5(b). Potential barriers around the isolated QD's induced by band bending could explain this decrease of the PL rise time with temperature, which is consistent with an increased rate of carrier transfer due to carriers having greater thermal energy to overcome these potential barriers around the QD's [13, 14]. Differences in the PL temperature dependence of InGaAs/GaAs and InAs/GaAs QD's (in which the effects of indium segregation and enrichment in the QD's and associated compositional fluctuations in the WL would be

minimized) may clarify the relative contributions from the temperature-dependent hole mobility and from potential fluctuations in the WL on carrier transfer to isolated quantum dots.

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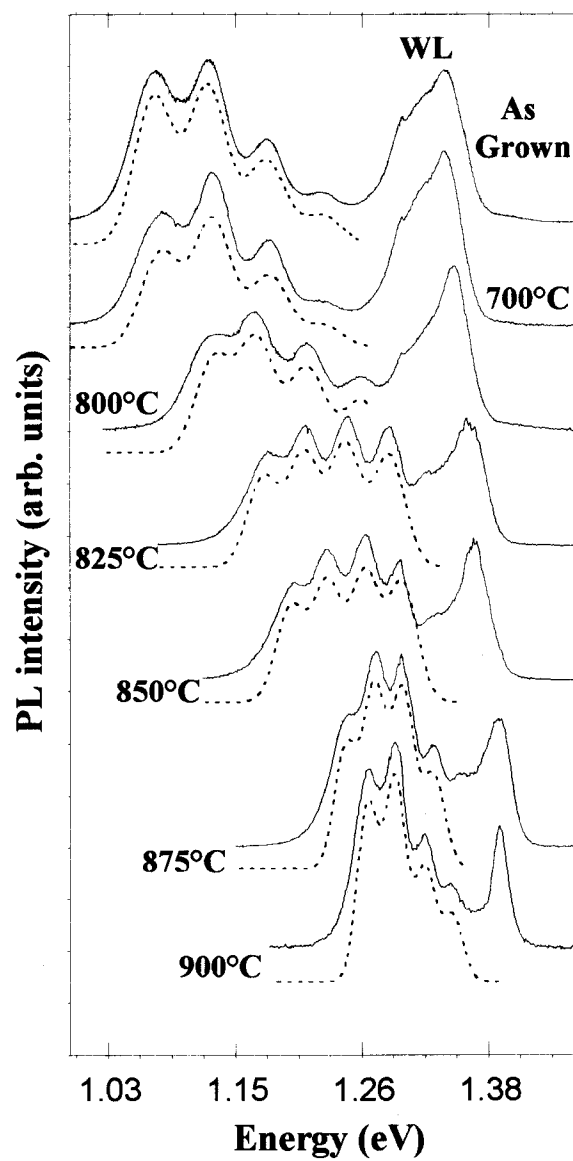


Figure 1. Effects of compositional intermixing of the InGaAs/GaAs interface on the radiative emission of QDs that show excited states luminescence. Anneal temperatures are shown. Anneal times were 30 seconds for all anneals. Dashed lines show the results of simulation by solution of the rate equations.

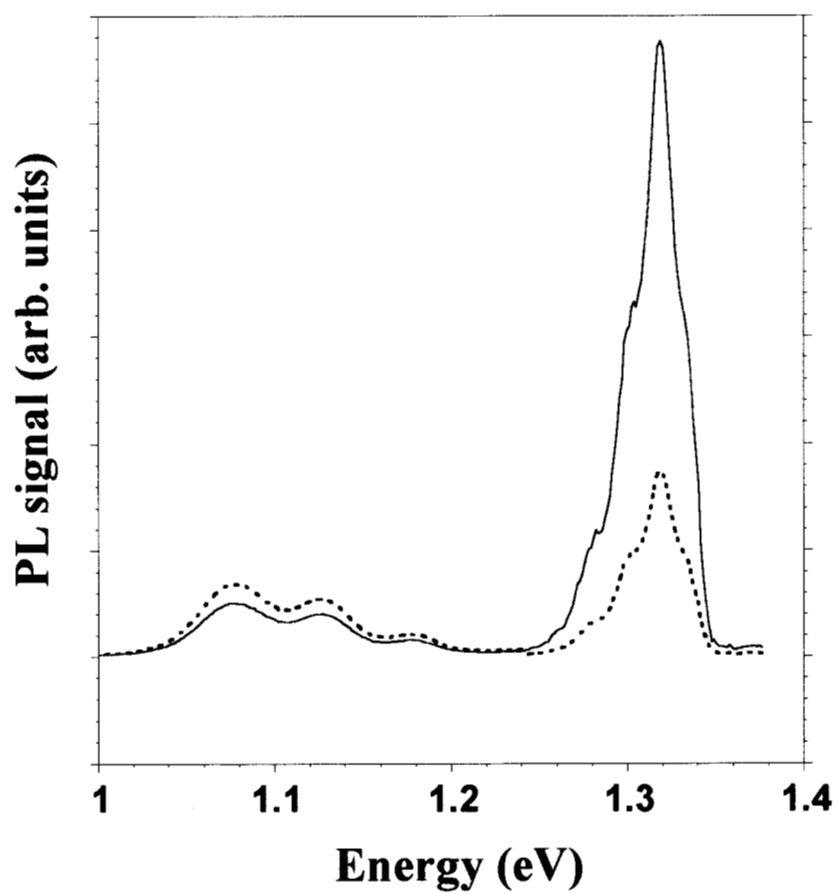


Figure 2. Comparison of initial (solid line) and post irradiation (dotted line) PL spectra (measured at 5 K) at a proton dose of $2.7 \times 10^{12}/\text{cm}^2$ from QD structures with low QD density (3.5×10^8 dots per cm^2). The spectra were obtained at constant excitation and show simultaneous emission from QD and wetting layer states.

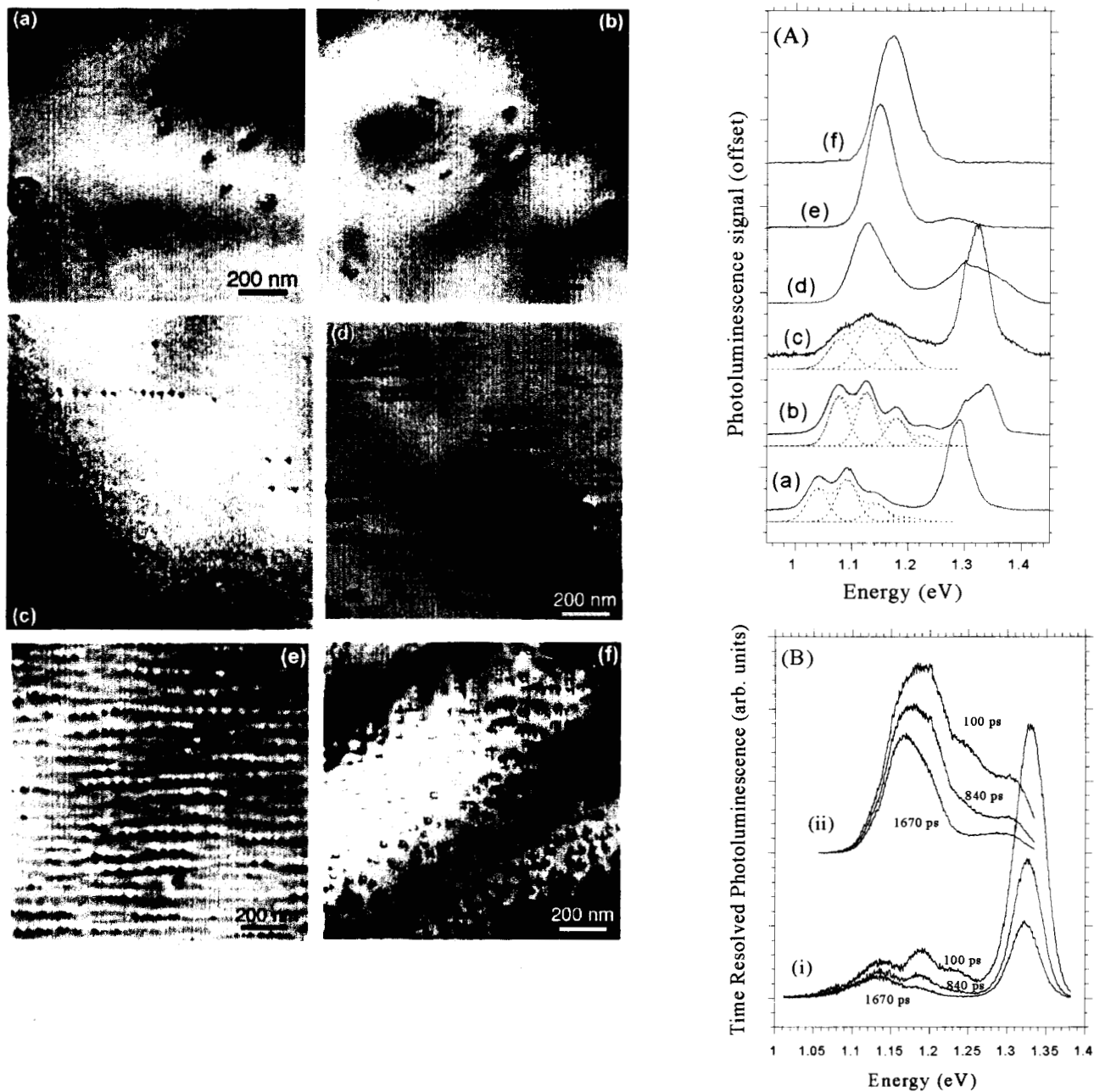


Figure 3. (Left) Representative images of varying concentrations and spatial arrangements in strained InGaAs/GaAs quantum dots obtained in simultaneous growth on different substrates with 100 nm capping layers. Imaging conditions were either off-zone-axis or axial bright field. (A) $\theta_m = 0.25 \pm 0.25$, (B) $\theta_m = 0.00 \pm 0.25$, (C) $\theta_m = 0.75 \pm 0.25$, (D) $\theta_m = 1.25 \pm 0.25$, (E) $\theta_m = 2.00 \pm 0.25$, and (F) $\theta_m = 0.00 \pm 0.25$, under growth conditions that give maximum island coverages. (a) PL spectra in InGaAs/GaAs quantum dots of varying concentrations. Spectra were taken at 77 K and are labeled as the corresponding plan view images in composite image on left. (b) Time-resolved PL spectra, integrated over a 50 ps temporal window with central time values of 100, 840 and 1670 ps after the excitation at 77 K for (i) sample (C), and (ii) sample (E).

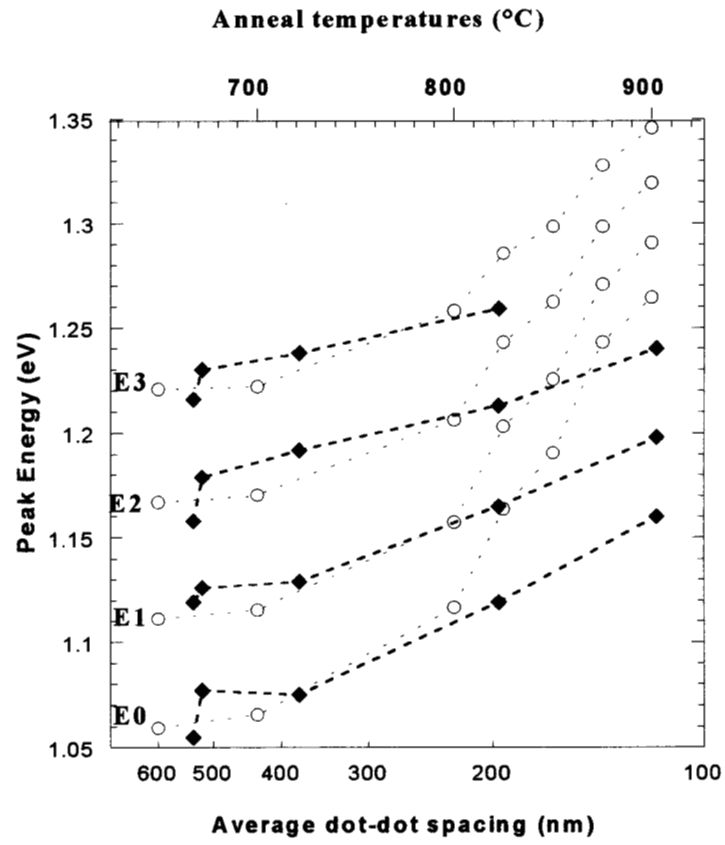


Fig. 4 (a) Level energies obtained from Gaussian fits of PL spectra (b) Variation of level energies obtained after post-growth annealing, which causes interdiffusion of the InGaAs/GaAs interface [6].

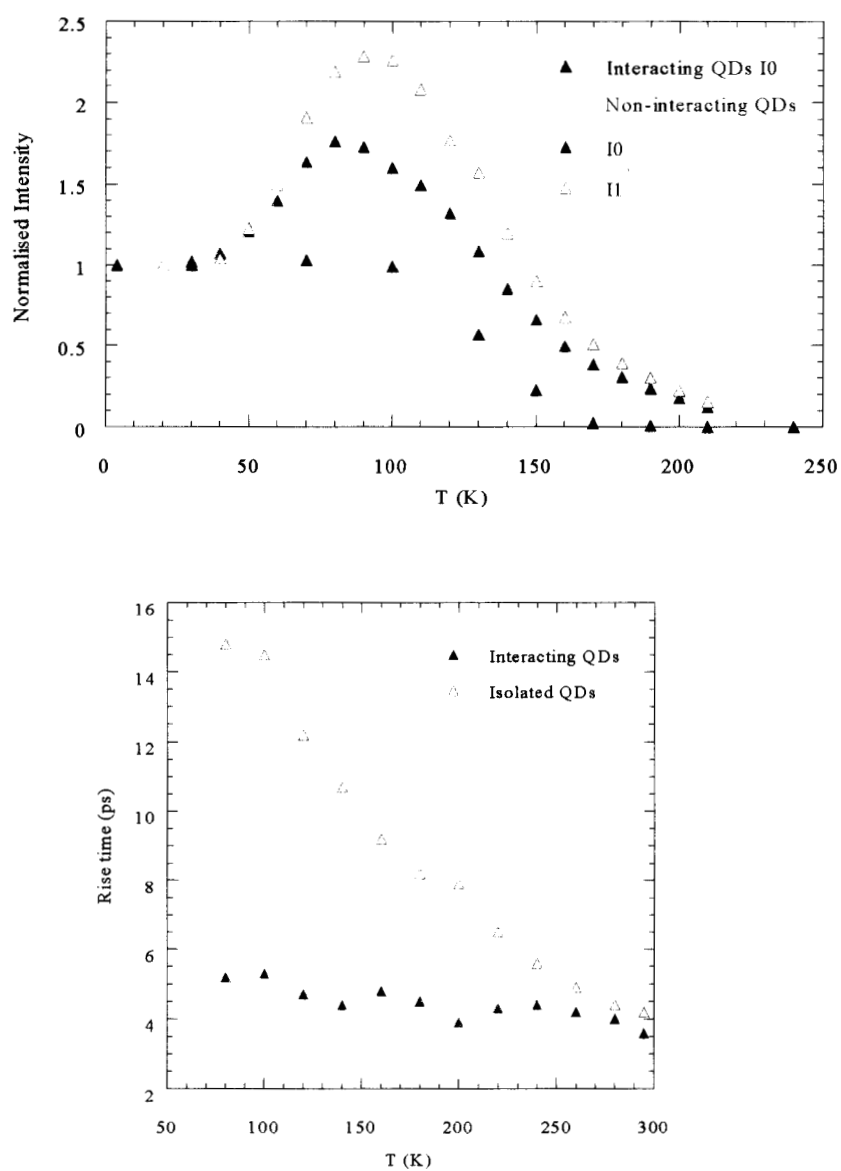


Figure 5. (a) Normalized integrated PL intensity as a function of temperature for interacting and isolated QD samples. Interacting QD samples are represented by filled blue triangles, isolated QDs are represented by filled (ground state) and hollow (first excited state) red triangles. (b) PL rise times as a function of excitation power for interacting and isolated QDs.